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14. ABSTRACT Equipment acquired and preliminary research results utilizing the equipment are reported. A synchronously-pumped optical parametric oscillator and a regenerative Ti:Sapphire amplifier system, both from Coherent, Inc., have been acquired and installed. The equipment has been used for optical characterization of metallo-dielectric photonic crystals (MDPCs) and for realization of electromagnetically-induced transparency (EIT) in semiconductor heterostructures. Ordered MDPCs have been successfully fabricated via colloidal assembly. EIT in semiconductors have been demonstrated for the first time in interband optical transitions in GaAs quantum well structures.

15. SUBJECT TERMS Electromagnetically-induced transparency, Metallo-dielectric photonic crystals, semiconductor heterostructures, colloidal self-assembly.

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Equipment

Regenerative Ti:Sapphire amplifier system (Wang Laboratory)

Vendor: Coherent, Inc.

System: Reg9000 amplifier pumped by a 10W Verdi laser

Price: \$160,000

Accessories: Laser wavemeter

Vendor: EXFO

Price: \$155,000

Optical parametric oscillator system (Deutsch Laboratory)

Vendor: Coherent Inc.

System: Synchronously-pumped optical parametric oscillator

Verdi V-10 pump laser

Price: \$148,000

Accessories:

Monochromator and liquid-nitrogen-cooled CCD chip detector

Vendor: Jobin Yvon

Price: \$31,200

Research

EIT AND SLOW LIGHT IN SEMICONDUCTORS (Wang Laboratory)

A variable optical delay line that can change the optical delay in ultrafast timescales is a critically sought after component in optical communication and optical signal processing. The controllable optical delay line can be used for optical data synchronization and can function as variable optical buffers. Recent advances in the investigation of electromagnetically induced transparency, a phenomenon that renders an opaque medium transparent through destructive quantum interference, have prompted the idea of using slow light induced by EIT in a semiconductor to realize compact and controllable optical delay lines.

With support from the Army Research Office and with equipment support from the Air Force Office of Scientific Research, we have carried out extensive studies on EIT in semiconductors. We have made the breakthrough of demonstrating for the first time EIT from an interband optical transition in semiconductors. We have shown that EIT can be realized in GaAs quantum well (QW) structures by using a variety of nonradiative quantum coherences including exciton spin coherence, biexciton coherence, and intervalence band coherence. Using EIT induced by a biexciton coherence, we have

achieved a 20-fold reduction in the absorption of an exciton resonance in a GaAs QW. While the above experimental studies were carried at low temperature, we are investigating the possibility of realizing EIT using the robust electron spin coherence which can persist to room temperature in semiconductors. Experimental studies aimed at a direct demonstrating of slow light are also currently under way.

METALLO-DIELECTRIC PHOTONIC CRYSTALS (Deutsch Laboratory)

Materials with high nonlinear conversion efficiencies are continually sought for nonlinear optical applications. We have developed a process for producing metallo-dielectric photonic crystals for this purpose. These structures are composed of sub-micron silica spheres which are first coated with nano-crystalline silver, and then self assembled into ordered solids. Nanometer-sized metal particles have been the focus of extensive studies for many years owing to their greatly amplified nonlinear optical (NLO) response, relative to that of bulk metals. These remarkable enhancements are attributed to enhancements of surface-induced electric fields at the plasma resonance of the nanoparticles, accounting for example for greatly enhanced second-harmonic-generation. We aim to combine the NLO properties of metallic nanocrystals with the novel optical response of photonic crystals (PCs), to form metallo-dielectric photonic crystals (MDPCs) - periodic composites of metal and dielectric. The nonlinear conversion efficiencies in these materials are expected to be exceptionally high, due to their enhanced surface area and unique photonic dispersion. New phase matching conditions should arise, leading for example to multi-directional frequency doubling.

We utilize colloidal self-assembly, which is a common method for fabricating three-dimensional PCs on optical length scales. Our unique method relies on fabrication of colloidal silica spheres coated with highly granular metallic nanoshells. These form the fundamental building blocks, later to be assembled into three-dimensional MDPCs. The optical properties of individual spheres and the MDPCs are controlled via sphere size and nanometal thickness. To obtain ordered MDPCs, self-assembly of the composite colloids is necessary. Normally metal-coated colloids do not form ordered structures, due to their large surface-roughness and strong interparticle forces which prevent ordering of metal-coated colloids. We find that when the spheres are coated in such a way that the metal shell is rough, yet on the average uniform in thickness, ordered single layers are readily formed. We believe that surface roughness may actually act to assist crystallization in these systems, by introducing local fluctuations in the electrostatic potential which may counter balance the van der Waals attraction between the spheres.

We have recently observed strong second harmonic and supercontinuum signals from disordered aggregates of silver-coated amorphous silica spheres. When illuminated with 100fs laser pulses of \sim 860nm, strong signals at 430nm were detected, in addition to a very broad and intense supercontinuum spanning the entire visible and NIR spectrum. Since silver does not generate second harmonics in the bulk due to its centrosymmetric crystalline structure, the frequency doubled signal originates only from the surfaces of the materials. By comparing to measurements on uncoated silica spheres we have so far

ascertained that the strong signals are due to the presence of silver in the system. We are currently conducting two separate studies to characterize the nature of these large signals. We aim to understand conversion efficiencies, dependence on core sphere size and ordering.

To obtain a thorough understanding of the optical response of nano-structured metallo-dielectrics, we are also conducting theoretical studies of these systems. Currently we are focusing on the properties of surface plasmon polaritons (SPPs) at metallic interfaces. We are seeking to understand the dispersive properties of SPPs on non-planar (curved) interfaces, as these pertain to the topology of the MDPCs. The immediate goal is to set practical limits on the applicability of metallo-dielectric nano-photonic and plasmonic systems, mainly by quantifying curvature guiding losses of SPPs. In the larger scheme, MDPCs pose a variety of questions concerning their optical modes which are still not well understood. In the future we aim to construct a comprehensive picture detailing a selection of these issues which are immediately relevant to our materials. Some important topics are: the nature of frequency conversion processes at microscopic spherical metal-dielectric interfaces; coupling of propagating SPP modes on spheres; nonlinear interactions between SPPs.

Publications

Mark Phillips and Hailin Wang, "*Spin coherence and electromagnetically induced transparency via exciton correlations*," Phys. Rev. Lett. **89**, 186401 (2002).

Mark Phillips and Hailin Wang, "*Electromagnetically induced transparency due to intervalence band coherence in semiconductors*," Opt. Lett., accepted for publication.

Mark Phillips, Hailin Wang, I. Rumyantsev, N.H. Kwong, R. Takayama, and R. Binder, "*Electromagnetically induced transparency via biexciton coherence*," submitted to Phys. Rev. Lett.

Phedon Palinginis and Hailin Wang, "*Vanishing and emerging absorption quantum beats from electron spin coherence in GaAs quantum wells*," Phys. Rev. Lett. (in press).

J. Bouwman, A. Randolph, D. Cassell, C. Rohde, M. Deutsch, "*Self Assembled Metallodielectric Photonic Crystals with Composite Silver/Silica Colloidal Spheres*," in preparation.

K. Hasegawa, J. Noeckel, M. Deutsch, "*Surface Plasmon Polariton Propagation around Bends at a Metal-Dielectric Interface*," accepted provisionally for publication in Appl. Phys. Lett., 2003.

Scientific personnel involved

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